Resonant Raman scattering by collective modes of the one-dimensional electron gas.

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We show that the low-energy peak in the polarized resonant Raman spectra of quantum wires, which is commonly associated with "single particle excitations", can be interpreted as signature of intra-band collective spin excitations. A broad maximum in the resonant depolarized spectra is predicted to exist above the frequency of the spin density excitation, due to simultaneous but independent propagation of spin- and charge-density modes.

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Inelastic light scattering by etched AlGaAs/GaAs-quantum wires at Helium temperatures shows pronounced features that are commonly interpreted in terms of inter- and intra-subband excitations of the interacting electron gas. Depending on the relative polarization of the incident and the scattered light, charge-density excitations (CDE, parallel polarization, polarized spectra) and spin-density excitations (SDE, crossed polarization, depolarized spectra) have been identified. Near resonance, when the frequency of the incident light is close to the frequency of the band gap, additional structures have been detected in both types of spectra which have been interpreted as being the signature of "single particle excitations" ("SPE") [1–4]. The intensities of these peaks depend strongly on the frequency of the incident light. Their physical origin is presently controversely discussed. In [1,2] where the electron density and the geometry of the wires have been such that only two subbands were occupied, the Fermi-liquid character of the 1D electron gas has been stressed. In [4], with several subbands involved in the inelastic scattering, the "SPE" have been interpreted as "energy-density fluctuations". Recent results of very elaborate RPA calculations [5] seem to be consistent with the absence of "SPE" at low excitation energies in 1D. By including two occupied subbands [6], the observed "SPE" peaks at low energies have been interpreted as an out-of-phase CDE in the two-band system in which the long-range part of the Coulomb interaction is partially cancelled [7]. However, as has been pointed out in [1], the energies of these excitations, as observed experimentally, seem to be much too small.

In this paper, we point out that the peaks at low excitation energy in the *polarized Raman spectra* which are strongly enhanced when the photon energy approaches the energy gap, can be understood within the theory of the collective excitations of the 1D electron gas with spin in the Luttinger approximation. We show that they are signatures of the collective SDE and appear at the same energy, namely $\hbar v_\sigma |q|$, as the SDE in the depolarized spectra (v_σ, q velocity and wave number of spin density excitations, respectively). They appear due to higher order terms in the Raman cross section related to the resonance condition. These terms induce a relaxation of the selection rules valid for non-resonant Raman scattering. Our results indicate for quantum wires a solution of the puzzle of the "SPE" that has been posed already more than two decades ago for inversion layers [8]. We quantify the suggestion of a singlet spin mode being responsible for the "SPE" peaks which has been mentioned to the best of our knowledge for the first time in [7].

We predict that, when $\hbar v_{\rm F} q/|E_{\rm G} - \hbar\omega_{\rm i}| \ll 1$ ($E_{\rm G}$ energy gap, $v_{\rm F}$ Fermi velocity), the strength of the "SPE" peak varies with the energy $\hbar\omega_{\rm i}$ of the incident light as

$$I_{\rm SPE} \propto |E_{\rm G} - \hbar\omega_{\rm i}|^{-4}\,, \tag{1}$$

in lowest order, and *increases* quadratically with temperature. As a further consequence of the resonance condition, we predict that structure associated with simultaneous propagation of spin- and charge density excitations should appear in the *depolarized spectra*. In contrast to the SDE in the polarized spectra, this is not a sharp peak but merely a rather broad maximum in the scattered intensity on the high energy side of the SDE-peak.

Our results show that the Raman excitation spectra of quantum wires at low energies, in the region of *intraband transitions*, can be understood within the non-Fermi liquid framework of the 1D electron gas.

Within the standard theory of Raman scattering [9] the differential cross section at $\omega = \omega_{\rm i} - \omega_{\rm f}$, the difference between the frequency of the light in the initial and final state, is given by the average

$$\frac{{\rm d}^2\sigma}{{\rm d}\Omega {\rm d}\omega} \propto \left\langle \sum_{\rm f} |M_{\rm fi}|^2 \delta(E_{\rm f} - E_{\rm i} - \hbar\omega) \right\rangle_{\rm i} \tag{2}$$

where i and f denote initial and final states, respectively, and $\langle\cdots\rangle_{\rm i}$ is the thermal average over the initial state. The transition matrix elements $M_{\rm fi}$ consist of terms proportional to $\boldsymbol{A}^2$ and $\boldsymbol{\Pi}\cdot\boldsymbol{A}$ ($\boldsymbol{A}$, $\boldsymbol{\Pi}$ vector potential and momentum operator, respectively). The latter has to be

treated in second order, and requires further approximations, especially near resonance. The final result, taking into account only one conduction and one valence band (with effective masses $m_{\rm c}$ and $m_{\rm v}$, respectively), can be written in terms of a generalized correlation function ($\boldsymbol{q} \equiv \boldsymbol{k}_{\rm i} - \boldsymbol{k}_{\rm f}$),

$$\frac{{\rm d}^2\sigma}{{\rm d}\Omega {\rm d}\omega} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_{\rm f}}{\omega_{\rm i}} \frac{n_\omega + 1}{\pi} \mathcal{I}{\rm m}\chi(\boldsymbol{q},\omega)\,, \qquad (3)$$

with $\chi(\boldsymbol{q},t) = {\rm i}\Theta(t)\langle\big[N^\dagger(\boldsymbol{q},t), N(\boldsymbol{q},0)\big]\rangle$, and the Bose distribution $n(\omega)$. The operator

$$N(\boldsymbol{q}) = \sum_{s,k} \frac{\gamma_s}{D(\boldsymbol{k})} c_s^\dagger(\boldsymbol{k}+\boldsymbol{q}) c_s(\boldsymbol{k}) \qquad (4)$$

contains the Fermion operators $c_s^\dagger(\boldsymbol{k})$, $c_s(\boldsymbol{k})$ with wave vector $\boldsymbol{k}$ and spin $s = \pm$. The coefficients are

$$\gamma_s = \gamma_0 \left(\boldsymbol{e}_{\rm i} \cdot \boldsymbol{e}_{\rm f} + {\rm i}s|\boldsymbol{e}_{\rm i} \times \boldsymbol{e}_{\rm f}|\right)\,. \qquad (5)$$

Here, $\boldsymbol{e}_{\rm i,f}$ are the polarization vectors of the incoming and outgoing electromagnetic fields, and

$$D(\boldsymbol{k}) = E_{\rm c}(\boldsymbol{k}+\boldsymbol{q}) - E_{\rm v}(\boldsymbol{k}+\boldsymbol{q}-\boldsymbol{k}_{\rm i}) - \hbar\omega_{\rm i} \qquad (6)$$

contains the energies of the valence and conduction bands $E_{\rm v}$ and $E_{\rm c}$ (in the effective mass approximation), respectively, and the wave vector of the incoming light, $\boldsymbol{k}_{\rm i}$. The prefactor γ_0 contains the matrix elements for the transitions between the valence and the conduction band. It is assumed to be constant in the following.

If $\hbar v_{\rm F} q \ll |E_{\rm G} - \hbar\omega_{\rm i}|$ and $\hbar\omega \ll |E_{\rm G} - \hbar\omega_{\rm i}|$, one can neglect the $\boldsymbol{k}$-dependence of $D(\boldsymbol{k}) \approx E_{\rm G} - \hbar\omega_{\rm i}$ (with $E_{\rm G} = E_{\rm G}^0 + \eta E_{\rm F}$ ($\eta = 1 + m_{\rm c}/m_{\rm v}$)). Here, $E_{\rm G}$ is the distance between the conduction and the valence band at the Fermi wave number $k_{\rm F}$ and $E_{\rm F} = \hbar^2 k_{\rm F}^2/2m_{\rm c}$. Then, the operator

$$N(\boldsymbol{q}) = \frac{\gamma_0}{E_{\rm G} - \hbar\omega_{\rm i}} \left[\boldsymbol{e}_{\rm i} \cdot \boldsymbol{e}_{\rm f}\, \rho(\boldsymbol{q}) + {\rm i}|\boldsymbol{e}_{\rm i} \times \boldsymbol{e}_{\rm f}|\, \sigma(\boldsymbol{q})\right] \qquad (7)$$

is proportional to the charge density $\rho(\boldsymbol{q}) = \rho_+(\boldsymbol{q}) + \rho_-(\boldsymbol{q})$ or to the spin density $\sigma(\boldsymbol{q}) = \rho_+(\boldsymbol{q}) - \rho_-(\boldsymbol{q})$ depending on whether incoming and outgoing light are polarized parallel or perpendicular, respectively. Thus, in lowest order, one observes charge-density excitations in polarized, and spin-density excitations in the depolarized configuration. This is the "classical" selection rule of Raman spectra of quantum wires and dots.

Close to the resonance, when $\hbar\omega_{\rm i} \approx E_{\rm G} + \hbar v_{\rm F} q$, the assumption of a constant energy denominator is no longer valid. We expect that the above selection rule is relaxed. This will now be shown for a quantum wire and expanding $D(\boldsymbol{k})^{-1}$ to first order in $\hbar v_{\rm F} q (E_{\rm G} - \hbar\omega_{\rm i})^{-2}$. Specifically, we assume $E_{\rm c} = \varepsilon_n + \hbar^2 k^2/2m_{\rm c}$, with the subband energies ε_n ($n = 0, 1, 2, 3\ldots$) determined by the confinement in the y- and the z-directions, and k the wave number for the x-direction. Assuming back scattering, $k_{\rm i} = q/2$, and considering only the lowest subband,

$$D(k) = E_{\rm G}^0 + \eta\frac{\hbar^2 k^2}{2m_{\rm c}} + \frac{\hbar^2 k q}{m_{\rm c}}\xi - \hbar\omega_{\rm i} + {\rm O}(q^2)\,, \qquad (8)$$

with $\xi = 1 + m_{\rm c}/2m_{\rm v}$. Since $q \ll k_{\rm F}$ we can linearize around $k = \pm k_{\rm F}$. Then, we can use the Luttinger model [11,12] in order to evaluate the correlation function $\langle N^\dagger(q,t) N(q,0)\rangle$. In this model, it is useful to introduce a decomposition of the energy spectrum into branches b that correspond to left- and right-moving excitations, $b = -$ and $b = +$, respectively and $N(q) = \sum_b N^{(b)}(q)$.

The expansion of the inverse of the energy denominator yields contributions to $N(q)$ which are of the form of the "energy density fluctuations" mentioned in [10],

$$\Delta N^{(b)}(q) = -\frac{b\eta\hbar v_{\rm F}}{(E_{\rm G} - \hbar\omega_{\rm i})^2} \sum_{s,k} \gamma_s \cdot (k - b k_{\rm F}) \times c_s^{(b)\dagger}(k+q) c_s^{(b)}(k)\,. \qquad (9)$$

These can be expressed by the above charge- and spin-density operators by using the bosonization technique developped earlier for the Luttinger model in [12,14].

After a straightforward calculation, one obtains terms of the form (7) but $\propto \hbar v_{\rm F} q/(E_{\rm G} - \hbar\omega_{\rm i})^2$ and additionally new contributions which are quadratic in the densities

$$\Delta N^{(b)}(q) = -\frac{\eta v_{\rm F}\gamma_0}{(E_{\rm G} - \hbar\omega_{\rm i})^2} \frac{\pi}{2L} \times \sum_k \Big[2{\rm i}|\boldsymbol{e}_{\rm i} \times \boldsymbol{e}_{\rm f}| \rho^{(b)}(k)\sigma^{(b)}(q-k) + (\boldsymbol{e}_{\rm i} \cdot \boldsymbol{e}_{\rm f}) : \rho^{(b)}(k)\rho^{(b)}(q-k) + \sigma^{(b)}(k)\sigma^{(b)}(q-k) :\Big]\,, \qquad (10)$$

where the $: \cdots :$ stand for the normally ordered product of the operators.

Equation (10) is the main result of this work. The evaluation of the corresponding correlation function can be done exactly but is considerably more complicated than for $N(q) \propto \sigma(q)$, for instance. However, the form of $\Delta N(q)$ shows that in general the spin density fluctuations will contribute to the cross section in the polarized configuration besides the charge density fluctuations. Correspondingly, signatures of the latter can be expected in the depolarized spectrum. The "classical" selection rule which says that charge-wave excitations appear only in the polarized configuration and spin-wave excitations only in the depolarized spectrum, respectively, is only valid in the lowest approximation, when the wave vector dependence of $D(k)$ is neglected.

For the evaluation of the contribution of the spin excitations to the correlation function of the polarized spectrum we need to calculate correlators of the form

$$\langle[\sigma(k,t)\sigma(-q-k,t), \sigma(k',0)\sigma(q-k',0)]\rangle\,, \qquad (11)$$

instead of the correlation function $\langle[\sigma(-q,t),\sigma(q,0)]\rangle$ which determines the cross section in the depolarized configuration, when the energy denominator is constant.

The calculation of the correlation functions can be performed by using the Luttinger Hamiltonian $H = H_\rho + H_\sigma$, where H_ρ is the quadratic form describing the charge density excitations [7,13,14] and the spin part is

$$H_\sigma = \frac{\pi\hbar v_{\rm F}}{L}\sum_{q>0}\left[\sigma^{(+)}(q)\sigma^{(+)}(-q)+\sigma^{(-)}(-q)\sigma^{(-)}(q)\right] -\frac{g_1}{L}\sum_{q>0}\sigma^{(+)}(q)\sigma^{(-)}(-q)\,. \qquad (12)$$

The Hamiltonian can be diagonalized by a Bogolubov transformation. The spectrum of the charge modes is

$$\omega_\rho(q) = v_{\rm F}|q|\left\{\left(1+\frac{g_1}{hv_{\rm F}}\right)\left[1-\frac{g_1}{hv_{\rm F}}+4\frac{V(q)}{hv_{\rm F}}\right]\right\}^{1/2}, \qquad (13)$$

with $V(q)$ the Fourier transform of the interaction potential and an interaction constant g_1 which describes a part of the exchange interaction. For the spin density excitations

$$\omega_\sigma(q) = v_{\rm F}\sqrt{1-\frac{g_1^2}{h^2v_{\rm F}^2}}\,|q| \equiv v_\sigma|q|\,. \qquad (14)$$

The result of the calculation of the correlation functions valid *near resonance* can be written in a closed form, but the remaining integrals have to be computed numerically [15]. For the present purpose, it is sufficient to consider $\hbar v_{\rm F}q << |E_{\rm G}-\hbar\omega_{\rm i}|$ which gives for the spin contribution to the polarized spectrum at the temperature T a peak at the frequency $v_\sigma q$,

$$\mathcal{I}{\rm m}\chi(q,\omega) = \frac{Lq\,(\eta\hbar v_\sigma\gamma_0)^2}{12\,(E_{\rm G}-\hbar\omega_{\rm i})^4}\left[\left(\frac{\pi k_{\rm B}T}{\hbar v_\sigma}\right)^2+\frac{q^2}{2}\right] \times\delta(\omega-v_\sigma q)\,, \qquad (15)$$

which corresponds to the same position in energy as that of the SDE peak in the depolarized spectrum *far from resonance*,

$$\mathcal{I}{\rm m}\chi(q,\omega) = \frac{Lq\gamma_0^2}{(E_{\rm G}-\hbar\omega_{\rm i})^2}\delta(\omega-v_\sigma q)\,. \qquad (16)$$

We note that although the peaks appear at the same energies, their strengths depend differently on the photon energy. While the weight of the SDE peak increases quadratically with increasing $|E_{\rm G}-\hbar\omega_{\rm i}|^{-1}$, the peak in the polarized spectrum increases with the 4th power. Also, the SDE-related peak in the polarized spectrum far from resonance is independent of the temperature, due to the linearization of the spectrum, while the peak in the polarized spectrum increases quadratically with T.

In the depolarized configuration, we obtain also a relaxation of the "classical" selection rules near resonance. The cross section in next higher order contains correlation functions of the form

$$\langle[\rho(k,t)\sigma(-q-k,t),\rho(k',0)\sigma(q-k',0)]\rangle\,. \qquad (17)$$

and no correlation functions with four charge density operators alone. Due to the absence of spin-charge coupling in the Hamiltonian, (17) factorizes into products of the type $\langle\sigma(-q-k,t)\sigma(q-k',0)\rangle\langle\rho(k,t)\rho(k',0)\rangle$ indicating independent motion of the spin and charge modes. Due to the presence of terms like (17), we do not expect structure in the cross section that is solely determined by the charge density excitations. Indeed, we find that the simultaneous propagations of the two types of excitations leads to a broad continuum in the depolarized spectrum above the frequency $\omega_\sigma(q)$.

Also in the contributions that are still higher order in $\hbar v_{\rm F}q(E_{\rm G}-\hbar\omega_{\rm i})^{-2}$, we do not find correlators that contain only charge density operators since the depolarized part of the cross section originates in the spin-orbit coupling ([9], cf. (5)) and the corresponding excitation processes are accompanied by spin-flip processes. This implies that all of the terms contributing towards the depolarized cross section must contain at least one pair of spin density operators, and structure related to the charge density excitations alone is absent.

Comparing with experiment, we first note that all works agree in the linear dependence of the excitation energy on the wave number of the peak associated with the "SPE" in the polarized spectrum. In [4], the velocity of the "SPE" has been found to be approximately the same as the velocity of the SDE determined from the depolarized spectra, and approximately equal to the Fermi velocity in the lowest occupied subband. Our results are consistent with this, if we assume that $g_1/hv_{\rm F} \ll 1$. If the Fermi velocity was determined independently, the spin interaction constant g_1 could in principle be determined. However, it is expected that g_1 is in any case very small [7] so that $v_\sigma = v_{\rm F}$ to a very good approximation. In [1], data have been presented (Figs. 1 and 2) which seem to indicate that "SPE" and SDE peaks are slightly different in energy, the velocity of the former being approximately $v_{\rm F}$ while that of the latter has been identified to be slightly smaller. However, when taking the error bars into account it is not possible to distinguish between the positions of the peaks. Thus, these results can also be considered to be consistent with our present model.

On the other hand, we cannot exclude that there are many-particle corrections to the spin excitations beyond our model. For instance, if the spin Hamiltonian (12) contained additional quartic terms, the poles of the above quartic correlators (11) would be different in energy from those of the correlators quadratic in the spin density. Also, corrections due to higher subbands could lead to

different velocities of low-energy excitations. However, the recent experiments on samples with several subbands occupied [4], show that this is very improbable.

To the best of our knowledge, there are up to now no systematic measurements of the dependence of the heights and the widths of the "SPE" peaks as functions of the photon energy and the temperature. Such measurements could provide further support for our interpretation. One should have in mind that the *precise* value of the gap energy $E_{\rm G}$ is not known. Measurement of the dependence on the photon energy of the incoming light would provide the possibility of determining $E_{\rm G}$. Closer to resonance, approximation (1) is insufficient. Here, one has to evaluate numerically the k-integral in the correlator without expanding $D(k)^{-1}$ [15].

Concerning the additional structure in the depolarized spectrum predicted above, we could find only very weak experimental evidence in Fig. 2 of [1]. These authors interprete a slight asymmetry in the peak associated with SDE as a signature of the "SPE". Our findings offer a different interpretation: the asymmetry could be due to the continuum contribution to the depolarized spectrum which originates in the motions of simultaneouly excited spin- and charge-density waves. However, further experiments using wires with only one subband occupied are necessary, in order to confirm or to disprove this interpretation.

Within the present model, we cannot comment on the experimental *interband* results in quantum wires with higher subbands involved, where "SPE" and SDE have clearly different excitation energies. Due to the comparatevely high excitation energies, the pecularities of the Luttinger model are absent in this region. Especially, one can expect the Fermi-liquid character of the electron gas to be restored. However, we also expect for these excitations corrections towards the Raman cross sections in both configurations due to wave vector dependent terms in $D(k)$. We suspect that these (i) do not obey the "classical" selection rules and (ii) will in general produce structures at different energies than those of SDE and CDE.

In summary, we have presented results for the intraband Raman spectra of a quantum wire with only one subband occupied. They are consistent with all of the experimental findings presently available at low excitation energies. We have shown that the low-energy "SPE" in the *polarized spectrum* near resonance can be interpreted as signature of the spin-density excitations of the 1D electron gas. When accepting this, the presently available data of resonant Raman scattering can be taken as indicating the charge-spin separation predicted by the Luttinger model and for the non-Fermi liquid character of the 1D electron gas at low excitation energies.

The measurement of the above predicted dependence of the peak intensities on the photon energy and on the temperature, namely $\propto |E_{\rm G} - \hbar\omega_{\rm i}|^{-4}$ and T^2, respectively, could further confirm our interpretation. In addition, we predict near resonance a continuum in the cross section which extends above the frequency of the spin excitations in the *depolarized spectrum*. It is related to simultaneous but independent propagation of spin and charge modes.

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